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EFFECT OF RESONANCE RADIATIVE PROCESSES ON THE AMPLIFICATI0N FACTOR

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The effect of a strong held in resonance with one of the transitions of a substance is to change the emission and absorption spectra of adjacent transitions [1-3]. These changes are due to three effects: variation of the common-level population, splitting of energy levels in a strong field, and nonlinear interference processes. The last effect can be interpreted as an interference and a modification of the frequency-correlation properties of step-by-step and two-photon processes in a strong field under resonance conditions [4, 5]. Undoubtedly, these effects should manifest themselves in the amplification (absorption) properties of a weak held at one transition in the presence of a strong field in resonance with an adjacent transition. Of particular interest are optical transitions which have a greater number of relaxation constants than microwave transitions. Any of the above-mentioned effects can be made to predominate by appropriate selection of transitions. Maximum change of the amplification (absorption) factor can be obtained in gases for uniformly broadened transitions. *It will be shown below that amplification can occur even it the lower level has a greater saturated population than the upper.*

We shall analyze the case when both fields are in resonance, as the change in amplification is then the greatest. To be specific, consider the scheme of transition with a common lower level shown in Fig. 1. The system is located in a strong field whose frequency ω is equal to the natural transition frequency ω_{mn} ($\omega = \omega_{mn}$). The amplification factor of a weak field at the center of the transition line gn can be found from

$$\alpha_\mu = -\hbar\omega_{gn}\left[c|E_\mu|^2/8\pi\right]^{-1} N2\mathrm{Re}\{iV_{gn}e^{i\omega_{gn}t}\rho_{ng}\}.$$

Here E_μ is the amplitude of the weak field at frequency $\omega_{mu} = \omega_{gn}$, N is the number of particles per unit volume, V_{gn} is a matrix element of the perturbation Hamiltonian for the weak field $V_{gn} = G_\mu \exp\{-i(\omega_{gn}t - \mathbf{k}_\mu \mathbf{r})\}$; $G_\mu = -\mathbf{E}_\mu \mathbf{d}_{gn}/2\hbar$, ρ_{gn} is a density matrix in the interaction representation. The system of equations for the density matrix has a standard form and becomes algebraic in the stationary case [3]. The particle velocity distribution considered in [3] was Maxwellian.

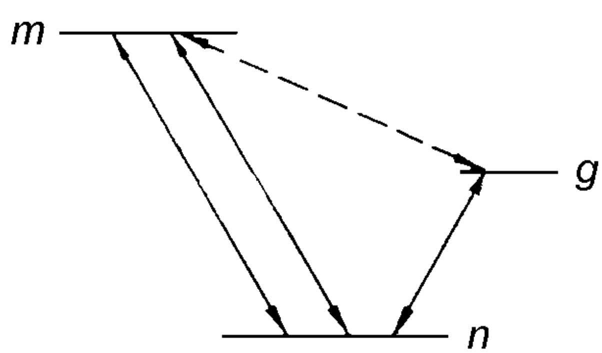

Figure 1:

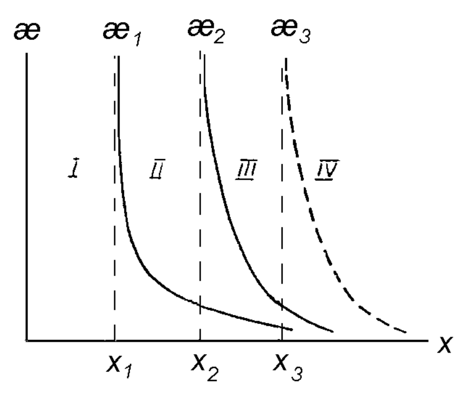

Figure 2:

Here we consider a monoenergetic particle beam with an arbitrary velocity v (this includes stationary particles). The change in amplification factor can be the greatest in such a case; on the other hand, this example makes it possible to find the interaction of the field with an individual atom. The monoenergetic beam

can be real and effective. An effective beam can be obtained by exciting atoms with a secondary monochromatic field onto one of the higher levels and letting them relax to the considered levels. A monochromatic field in a gas with nonuniform spectral line broadening interacts only with atoms the velocity projection of which in the $\Delta v_0 \approx 2\gamma_0/k_0$ interval is near the "resonance" velocity with a projection v_0 in the $\mathbf{k}_0$ direction. Here k_0 is the absolute value of the wave vector of the secondary excitation field, and $2\gamma_0$ is the line-width for an individual atom at the excitation transition. It is thus possible to ensure that the atoms have a negligible velocity spread on levels with which the nonlinear interference processes are associated. To this case also belong transitions for which the sum of the collision and natural widths is comparable with the Doppler width, for example, the $3s_2 - 3p_4$ transition in neon.

In a monoenergetic beam ω_{gn} and ω_{mn} should be understood as the Doppler-shifted natural frequencies of the corresponding transitions. In this case, the amplification factor α_μ at the center of the line and the population difference $N(n_g - \rho_{nn})$ saturated by the strong field are respectively

$$n_g - \rho_{nn} = \Delta n_{gn} - \Delta n_{mn}\left(1 - \frac{\gamma_{mn}}{\Gamma_m}\right)\frac{2\Gamma}{\Gamma_n}\frac{|G|^2}{\Gamma^2(1+\varkappa)}, \tag{1}$$

$$\frac{\alpha_\mu}{\alpha_\mu^0} = \frac{\Gamma_{gn}}{\Gamma_{gn} + |G|^2\Gamma_{gm}^{-1}}\left\{1 - \frac{\Delta n_{mn}}{\Delta n_{gn}}\frac{|G|^2}{\Gamma^2(1+\varkappa)}\left[\left(1 - \frac{\gamma_{mn}}{\Gamma_m}\right)\frac{2\Gamma}{\Gamma_n} + \frac{\Gamma}{\Gamma_{gm}}\right]\right\}. \tag{2}$$

Here $N\Delta n_{ik} = N(n_i - n_k)$ are unsaturated population differences at the respective transitions (for $|G|^2 = 0$); N is the particle concentration at the ground state; G is a matrix element of the perturbation Hamiltonian of the strong field: $G = -\mathbf{E}\mathbf{d}_{\mathbf{mn}}/2\hbar$; $\hbar\Gamma_i$ are the energy level widths; Γ_{ik} ($\Gamma_{mn} = \Gamma$) usually appear as half- widths of the corresponding transition lines (in angular frequencies); and γ_{mn} is the probability of relaxation transition from level m to lever n per unit time. The saturation parameter $\varkappa$ is given by

$$\varkappa = (\Gamma_m + \Gamma_n - \gamma_{mn})(\Gamma_m\Gamma_n\Gamma)^{-1}2|G|^2 = \tau^2 2|G|^2,$$

and α_μ^0 is the amplification factor at the line center without the strong field. The term proportional to $|G|^2$ in the denominator of the first factor in (2) reflects the effect of level splitting in the strong field, while the term inversely proportional to Γ_{gm} accounts for the presence of frequencv-correlated transitions between the states m and g via an intermediate level (nonlinear interference processes) [4, 5].

From (1) follows that an inversion of the sign of $n_g - \rho_{nn}$ should take place if the signs of Δn_{gn} and Δn_{mn} are the same and if the field obeys the relation

$$\left(1 - \frac{\gamma_{mn}}{\Gamma_m}\right)\frac{2|G|^2/\Gamma\Gamma_n}{1+\varkappa} > \frac{\Delta n_{gn}}{\Delta n_{mn}} \tag{3}$$

However, the contribution of interference effects causes α_μ sign inversion to take place at lower field values provided the inequality

$$\left[1 - \frac{\gamma_{mn}}{\Gamma_m} + \frac{\Gamma_n}{2\Gamma_{gm}}\right]\frac{2|G|^2/\Gamma\Gamma_n}{1+\varkappa} > \frac{\Delta n_{gn}}{\Delta n_{mn}} \tag{4}$$

is satisfied.

Comparing (3) and (4) we observe that the difference between the critical fields in the first and second case is the greater the stronger the inequality

$$\Gamma_{gm} \leq \Gamma_n/2\left(1 - \frac{\gamma_{mn}}{\Gamma_m}\right). \tag{5}$$

In particular, if $\Gamma_m = \gamma_{mn}$ the external field has no effect on the population of level n and the change of the sign of α_μ is due solely to interference effects. If inequality (3) is observed and (5) is not, the absolute value of the inverted population difference $|n_g - \rho_{nn}(\varkappa)|$ will for some fields exceed the interference term $\Delta n_{mn}|G|^2/\Gamma_{gm}\Gamma(1+\varkappa)$ and cancel its effect.

Figure 2 shows schematically the analysis of the conditions of inversion of α_μ sign at the line center provided Δn_{nm} and Δn_{gn} have the same signs. The values of $\varkappa$ and $x = \Delta n_{mn}/\Delta n_{gn}$ are plotted along the Y and X axes respectively. The curve $\varkappa_1(x)$ describes fields for which α_μ turns into zero. In domain I no sign inversion can take place whatever the field. *In domain II sign inversion takes place without a change of the sign of the population difference $n_g - \rho_{nn}(\varkappa)$.* The curve $\varkappa_2(x)$ describes fields for which this difference is zero. In domain III the population difference $n_g - \rho_{nn}(\varkappa)$ changes its sign. In domain IV the interference contribution is counterbalanced by the population difference in case condition (5) is not satisfied.

The expressions for curves $\varkappa_1$, $\varkappa_2$, $\varkappa_3$ and for the critical population difference ratios are

$$\varkappa_i(x) = (x x_i^{-1} - 1)^{-1}; \quad x_2 = \frac{\Gamma_m - \gamma_{mn} + \Gamma_n}{\Gamma_m - \gamma_{mn}};$$
$$x_{1,3} = \frac{\Gamma_m - \gamma_{mn} + \Gamma_n}{\Gamma_m - \gamma_{mn} \pm \Gamma_n \Gamma_m (2\Gamma_{gm})^{-1}}. \tag{6}$$

The fact that amplification is possible when $x < x_2$, $\varkappa < \varkappa_2$, even if $n_g - \rho_{nn}(\varkappa) < 0$ can be explained as follows. For relatively low external fields the nonlinear interference effects can be considered as a contribution of two-photon transitions $g \leftrightarrow m$ via an intermediate level n to the emission (absorption) of $\hbar\omega_\mu$ quanta (4). For amplification to take place at the frequency ω_μ it is sufficient that $n_g > \rho_{mm}$ irrespective of the sign of $n_g - \rho_{nn}$ (so that always $x_i > 1$). The range of x values, x_2 to x_1, for which *inversion of the α_μ sign can take place without inversion of $n_g - \rho_{nn}$* is the greater the stronger the inequality (5).

In solids, in which linewidths are as a rule considerably broader than levels, condition (5) can be satisfied only when $\Gamma_m \approx \gamma_{mn}$. If this condition is satisfied, the strong and weak fields interact mainly through interference processes and

$$x_1 \approx 2\Gamma_{gm}/\Gamma_m, \quad x_2 >> x_1. \tag{7}$$

In purely spontaneous relaxation in gases, when $\Gamma_{ik} \approx (1/2)(\Gamma_i + \Gamma_k)$, expression (7) turns into

$$x_1 = 1 + \Gamma_g/\Gamma_m, \quad x_1 \to 1, \quad \Gamma_g/\Gamma_m \to 0.$$

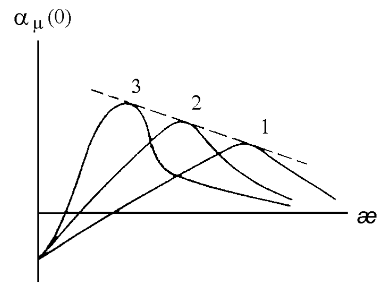

Figure 3: $\alpha_\mu(0)$ as a function of $\varkappa$. $x = \Delta n_{mn}/\Delta n_{gn} > 0$ increases from curve 1 to curve 3 ($x^{(1)} < x^{(2)} < x^{(3)}$).

In the other limiting case ($\gamma_{mn} << \Gamma_m$) and for purely spontaneous relaxation in gases we have

$$x_1 = 1 + \frac{\Gamma_n \Gamma_g}{\Gamma_m(\Gamma_m + \Gamma_n + \Gamma_g)}; \quad x_2 = 1 + \frac{\Gamma_n}{\Gamma_m};$$
$$x_3 = 1 + \frac{\Gamma_n(2\Gamma_m + \Gamma_g)}{\Gamma_m(\Gamma_m + \Gamma_g - \Gamma_n)} \quad (\Gamma_m + \Gamma_g > \Gamma_n).$$

Thus the most favorable relationship in this is $\Gamma_n >> \Gamma_m >> \Gamma_g$. At the same time, interference process are effective everywhere to the right of the curve $\varkappa_1(x)$, and a slight excess of level g population over level n population is sufficient *for amplification to take place in a strong field E even if $n_g - \rho_{nn}(\varkappa) < 0$.*

If n is the ground level, it is necessary to pass to the limits as follows

$$\Gamma_n \to 0, \quad \Gamma_m - \gamma_{mn} \to 0, \quad (\Gamma_m - \gamma_{mn})/\Gamma_n \to 1.$$

From (6) we have

$$x_1 = 1 + \frac{2\Gamma_{gm} - \Gamma_m}{2\Gamma_{gm} + \Gamma_m}, \quad x_2 = 2. \tag{6'}$$

Thus, in solids the value of x_1 in the given case is nearly two and differs little from the value of x_2.

For gas and purely spontaneous relaxation (6') gives $x_1 > 1$ if $\Gamma_m >> \Gamma_g$. In this case with $n_m = 0$ a slight additional excitation of the g level is sufficient for the strong field ($\varkappa >> 1$) to produce amplification.

From the above discussion follows that the contribution of nonlinear interference processes to amplification is most significant under the following conditions. In the considered transition scheme, the common level should be the broadest, the final level of the two-photon transition should be broader than the starting level, and the relaxation of the upper strong-field transition level should take place mainly by decay onto the lower level of this transition.

Let us evaluate the effect of nonlinear interference processes in the interaction of the transitions $3s_2 - 2p_4$ and $2s_2 - 2p_4$ in neon. The following relaxation constants are used $\Gamma_m = 3 \cdot 10^7 \mathrm{sec}^{-1}$, $\Gamma_n = 5 \cdot 10^7 \mathrm{sec}^{-1}$, $\Gamma_g = 10^7 \mathrm{sec}^{-1}$, $\gamma_{mn} = \gamma_{gn} = 0.5 \cdot 10^7 \mathrm{sec}^{-1}$. Thus, $x_1 \approx 1.2$ and $x_2 \approx 3$ if the strong-field transition is $3s_2 - 2p$, and $x_1 \approx 3.1$ and $x_2 \approx 11$ otherwise. In the first case $(x_2 - x_1)/x_1 \approx 150\%$, while in the second $(x_2 - x_1)/x_1 \approx 255\%$. In both cases (5) is satisfied as an inequality.

Let us now analyze the effect of the strong field, given by the common denominator in (2), on the amplification factor at the center of the line. This term reflects the splitting of energy levels m and n under the effect of the

strong field. Since $|G|^2$ enters both the numerator and denominator of (2) with different weights, the absolute value of the factor α_μ at the line center first increases and then falls with increasing intensity of the external field $\varkappa > \varkappa_1$ for fixed $x > x_1$.

Thus, for any $x > x_i$ there is an optimum external field at which the factor α_μ at the line center is maximum. The optimum field $\varkappa_{opt}$ as a function of the ratio $x = \Delta n_{mn}/\Delta n_{gn}$ is given by

$$\varkappa_{opt}(x) = \varkappa_1(x)\{1 + \sqrt{1 + [x_1\varkappa_1(x)]^{-1}(2\tau^2\Gamma_{gm}\Gamma_{gn}x + x_1)}\}, \quad x > x_1. \tag{8}$$

Here x_1 and $\varkappa_1(x)$ are given by (6). With increasing x ($x > x_1$) the optimum field decreases at the limit as $x_1/x\{1 + \sqrt{1 + 2\tau^2\Gamma_{gm}\Gamma_{gn}}\}$. For any fixed field, α_μ depends linearly on x. The dependence of $\alpha_\mu(0)$ on $\varkappa$ and x is qualitatively illustrated in Fig. 3. For example, for the neon transitions considered above (strong-field transition $3s_2 - 2p_4$) $x = 4.14$ corresponds to an optimum field $\varkappa_{opt} = 2$. Estimates indicate that for these optimum values of $\varkappa$ and x, the change in α_μ accompanied by sign inversion is quite large $\alpha_\mu/\alpha_\mu^0 = -32$, and the optimum is sharply pronounced. Thus, for $\varkappa$ equal to one half of its optimum value the absolute value of α_μ decreases by a factor of nearly three. For $\varkappa$ 50% greater than $\varkappa_{opt}$, $|\alpha_\mu|$ drops by a factor of 60.

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